UCRL- 91410
PREPRINT

Comparative Concentrations of ^{137}Cs, ^{90}Sr, $^{239,240}Pu$ and ^{241}Am in Tissues of Fish from the Marshall Islands and Calculated Dose Commitments from their Consumption

V.E. Noshkin
K.M. Wong
R.J. Eagle
W.L. Robison

February 1985

Lawrence Livermore National Laboratory

DISCLAIMER

Abstract

Body burdens of ^{90}Sr, ^{137}Cs, and the transuranics in bottom feeding fish from Marshall Island Atolls are derived, in part, from the quantities of the radionuclides irreversibly fixed to ingested carbonate material. Radionuclide concentration factors for different species of fish are characterized by relating tissue concentrations to those in filtered seawater. For bottom feeding fish, the values are lower at the lesser contaminated atolls than those values determined for the same species at the more contaminated atolls. These fish have the ability to lower their gut pH during feeding. When this occurs, there is a dissolution of a fraction of the ingested calcium carbonate containing radionuclides that were fixed or fused internally to the material during nuclear testing. Fractions of the radionuclides released during solution of the carbonate matrix are available for passage across the gut wall. Amounts released to solution in the gut are proportional to the levels of contamination at the different atolls. Concentration factors for higher trophic level species, which do not rely on sediments or coral for their source of food, show no such trends between differently contaminated atolls. A two-source model used to compute the internal concentrations is described. The model assumes radionuclides are derived from the "equilibrium labelled environment" and the sedimentary "bound" source term.

^{241}Am seems to be more biologically available than $^{239,240}Pu$ to higher trophic level species from the lagoons, whereas at lower trophic levels the opposite seems to be the case. ^{137}Cs is presently the largest contributor to the small radiological dose to man from the marine fish pathway, with the transuranics contributing from 2-30% of the total dose. There is little

reason to single out the transuranics as potential hazards in the marine fish ingestion pathway at these atolls, unless parts of the fish that are not normally consumed by man are included in the human diet.

Introduction

Results from our transuranic research program at the Pacific Proving Grounds, described in the first DOE Symposium Volume (Noshkin, 1980), focused primarily on the geochemical behavior of plutonium in the marine environment of Enewetak Atoll. In subsequent years information on plutonium oxidation states, the relative behavior of $^{239,240}Pu$ and ^{241}Am in both marine environments of Enewetak and Bikini, mobilization of plutonium and americium from lagoon sediments to seawater, and other geochemical topics was compiled and assembled for publication (Noshkin and Wong, 1980; Noshkin et al., 1984a). Concurrent with these studies, several biological investigations were initiated in an attempt to link the sedimentary source terms and geochemical observations with biological accumulation. There was also a need for related work to provide an updated estimate of potential radiological dose to individuals via the marine food chain at Enewetak, Bikini, and at other Marshall Island Atolls; fish is the major marine food product at these atolls. Detailed studies on transuranics and other radionuclides accumulated by these organisms were therefore needed.

Studies began at Enewetak Atoll in 1976 and were expanded to include sampling and analyses of fish from Bikini Atoll in 1977. In 1978, a radiological study at several Northern Marshall Atolls was undertaken. The northern atolls were contaminated to different degrees by intermediate range fallout resulting primarily from the detonation of a thermonuclear device, Bravo, in the 1954 Castle series of tests at Bikini. Fish and other samples were collected for analyses at the atolls of Rongelap, Rongerik, Likiep, Taka, Wotho, Ailinginae, Bikar, Ailuk, Ujelang, and the islands of Mejit and Jemo. In the early 1980s, we extended our investigations to include studies at Johnston Atoll, where plutonium in the marine environment was derived from global fallout deposition and local deposition from nonnuclear events. As a consequence of three THOR missle aborts, which occurred at Johnston Atoll in 1962 during the atmospheric nuclear testing program conducted by the United States, quantities of nonfissioned transuranics were scattered over the atoll and the surrounding marine environment.

Analyses of the fish collected from the Northern Marshalls in 1978 generated considerable data . Some of the results have been discussed elsewhere (Noshkin et al., 1981; Robison et al., 1981a), but a significant fraction of the data has not been assembled previously for discussion. In this report we discuss the results for ^{90}Sr, ^{137}Cs, ^{239,240}Pu, and ^{241}Am in a variety of fish representing marine trophic levels II to V. Results for some radionuclides in fish from Bikini, Enewetak, and Johnson will be published in a separate report (Noshkin et al., 1984b), since the data base is extensive and the accumulation of transuranics by fish from the environmental sources at these atolls requires additional study. We relate regularities and differences in concentrations with respect to tissue distributions, trophic level, and feeding habits. Differences in the values of concentration factors for different species are emphasized. Finally, to demonstrate the relative significance between transuranics, ^{137}Cs, and ^{90}Sr in dose-to-man estimates from a marine fish food pathway, we calculate radiological doses, basing them on the average radionuclide concentrations in fish muscle tissue and assuming an average daily intake of 200 g.

General Information

Collections and Methods

Throw nets were used exclusively to catch reef fish from the locations indicated by letter and number designators shown in Fig. 1. We used this identification system rather than the Marshallese island name for convenience only. The areas fished were close to shore in water not more than a few feet deep. Water and surface sediment was sampled at most locations in anticipation of relating tissue concentrations to environmental levels. Large pelagic and benthic fish were collected on sport fishing gear while trolling in the lagoon. All fish were returned to the ship, segregated by species, placed in plastic bags, and frozen in freezer units. The samples were shipped to Lawrence Livermore National Laboratory (LLNL), Livermore, California, for processing.

Species Collected, Feeding Habits, and Trophic Level Relationships

The principal species collected are commonly eaten by the Marshallese. They are relatively abundant; have different feeding habits; and for certain species, such as the surgeonfish and mullet, represent fish that were excellent indicator species, having the highest transuranic concentrations in edible muscle tissue. It was not always possible, however, to obtain an adequate number of the same species at every location sampled because of tides, insufficient time, and depletion due to overfishing at inhabited atolls. Feeding habits and trophic level assignments listed below are from descriptions by Hiatt and Strasburg (1965).

Various reef fish were collected. Mullet, <u>Crenimugil crenilabis</u> and <u>Neomyxus chaptalii</u>, are herbivorous, detrital feeders that ingest considerable quantities of bottom sediment along with food. Convict surgeonfish, <u>Acanthurus triostegus</u>, are herbivorous browsers that feed on small algal fronds and filamentous algae that grow on reef rock or on the base of dead coral. The unicornfish, <u>Naso lituratus</u>, also a herbivore, browses on larger seaweed growing in sandy and rocky areas. Rabbitfish, <u>Siganus rostratus</u>, are herbivorous browsers, but they will occasionally feed on fleshy items found in garbage dump areas. Rudderfish, <u>Kyphosus cinerascens</u>, are strictly herbivorous browsers. All of the above fish belong to the second trophic level. Goatfish, <u>Mulloidichthys samoensis</u>, consume fossorial and other benthic fauna, including small clams, crustaceans, other invertebrates, and small fish. This species belongs to the third trophic level. Threadfin, <u>Polydactylus sexfilis</u>, and flagtail, <u>Kuhlia taeniura</u>, feed on benthic fauna and also belong to the third trophic level. Parrotfish, <u>Scarus sordidus</u>, are common reef-dwelling, grazing omnivores feeding on live coral heads and occasional algae. Parrotfish are placed in the fourth trophic level because their food source (live coral polyps which feed on zoo plankton) are assigned to the third trophic level.

Larger benthic, midwater, and surface carnivores were also occasionally collected from lagoons. Grouper, <u>Epinephelus sp.</u>, are benthic carnivores of the third trophic level that feed on small fish and invertebrates. Jacks, <u>Caranx melampygus</u>, and <u>Elegatis bipinnulatus</u>, rainbow runner, are fast-swimming carnivores that feed on small fish and squid. <u>Elegatis bipinnulatus</u> may occasionally eat swimming crustacea. Snappers, <u>Aprion virescens</u> (grey snapper) and <u>Lutjanus bohar</u> (red snapper) are hovering, midwater-to-surface carnivores. Another snapper, <u>Letherinus kallopterus</u> (pigfish) is a bottom dweller that feeds primarily on benthic crustacea. Jacks and snappers are in the fourth trophic level. Tuna, <u>Euthynnus affinis</u> (bonito), <u>Thunnus</u>

albacares, and Gymnosarda nuda, and mackerel, Grammatorcynus billineatus, are large, rapid-swimming carnivores that feed on small fish and any other prey of proper size. They represent species of the fifth trophic level. In the remainder of this report, common rather than scientific names will be used for convenience.

Sample Processing and Analysis

The fish from each location were counted and partially thawed. The total weight, length, and sex of each fish was recorded. Each fish was dissected into muscle tissue, bone (cranial and thoracic, vertebrae and ribs, and pelvic and pectoral girdle), skin and scales (fins discarded), stomach contents, liver, and remaining viscera that included large and small intestines with contents, stomach wall, spleen, kidney, and mesenteries. The concentrations determined in the viscera samples are regrettably less descriptive than those for other tissues because of the matrix of organs and tissues represented. In some instances, however (in conjuction with our Bikini and Enewetak studies), a finer division of the visceral components was made for analyses. Each separate tissue and organ of the species from the same catch was pooled. It was necessary to pool tissues from a particular catch for the analysis of the low concentrations of transuranics anticipated in edible flesh. This resulted in the mixing of several populations (weight classes) and fish of different sex. Since mixing masked any differences in concentration related to weight (size), sorting of different size classes for processing was accomplished, in some instances, to assess any relationship of concentration to weight. We were unable to relate any differences in concentrations of specific radionuclides with sex. Gills were separated from the fish but not analyzed.

Our experience prior to 1978 showed that gills were frequently contaminated with sediment. Since gills are not eaten, little academic information would be gained from their analysis because of the possible contamination.

After the wet weight was determined, each pooled fish tissue sample was dried in ovens at 90°C to constant dry weight and dry ashed in muffle furnaces at 450°C for approximately 72 h. We processed 2625 fish on this program to make up the pooled number of samples of different species shown in Table 1. Shown also in Table 1 are the mean dry/wet tissue weights. The dry/wet stomach content values are of particular interest, since the differences noted attest to the different feeding habits of the species. The consistency of this ratio among each species from all atolls shows that each species is probably feeding on similar material at all atolls. The wet:dry quotient for the stomach content of mullet and parrotfish was similar to that for carbonate sedimentary material.

Samples were transferred to aluminum containers, sealed, and analyzed by gamma spectrometry. Gamma-spectrometry measurements were made on all separated samples at LLNL using a variety of Ge (Li)-diode detector systems. Counting times were usually 1000 min or longer for each sample. Except at Bikini and Enewetak, the only radionuclides, other than naturally occurring ^{40}K, detected in fish muscle tissue by gamma spectrometry were ^{137}Cs and occasionally ^{60}Co.

After gamma analysis the samples were either sent to a contractor laboratory or retained at LLNL for radiochemical separations of ^{90}Sr, ^{137}Cs, $^{238,239,240}Pu$ and ^{241}Am. These nuclides were judged to be of potential significance for dose assessments. The ^{137}Cs was radiochemically separated from muscle tissue and analyzed to confirm the measurements made by gamma spectrometry, which in turn provided a useful interlaboratory calibration for quality control. At LLNL, plutonium and americium were separated from the

TABLE 1 Dry/wet Tissue Weights

Fish common name	# of pooled samples	Atolls sampled[a]	Muscle	Bone	Stomach contents	Viscera	Skin	Liver
Reef fish								
Trophic Level II								
Surgeonfish	31	A,C,D,S,L,F, G,H,J,I,M	0.22 ± 0.01	0.59 ± 0.03	0.15 ± 0.04	0.19 ± 0.04	0.38 ± 0.03	0.23 ± 0.03
Crenimugil	16	A,C,D,L,F,M	0.23 ± 0.01	0.60 ± 0.05	0.62 ± 0.05	0.35 ± 0.07	0.53 ± 0.05	0.24 ± 0.05
Neomyxus	11	A,D,F,G,H,M	0.23 ± 0.01	0.58 ± 0.03	0.57 ± 0.03	0.41 ± 0.06	0.51 ± 0.03	0.24 ± 0.03
Rabbitfish	1	L	0.23	0.53	0.12	0.14	0.30	0.22
Rudderfish	1	L	0.21	0.48	0.07	0.14	0.45	0.23
Unicornfish	1	S	0.22	0.59	0.13	0.14	0.57	0.16
Trophic Level III								
Goatfish	19	A,C,L,F,G, J,I,M	0.23 ± 0.02	0.52 ± 0.05	0.22 ± 0.08	0.29 ± 0.05	0.50 ± 0.05	0.25 ± 0.03
Threadfin	3	S,G,I	0.24 ± 0.03	0.59 ± 0.02	0.17 ± 0.03	0.24 ± 0.05	0.47 ± 0.01	0.26 ± 0.03
Flagtail	2	R,I	0.24 ± 0.01	0.63 ± 0.05	0.17 ± 0.02	0.29 ± 0.06	0.61 ± 0.03	0.27 ± 0.05
Trophic Level IV								
Parrotfish	10	C,D,L,F,G,I,M	0.22 ± 0.04	0.56 ± 0.02	0.44 ± 0.09	0.41 ± 0.04	0.43 ± 0.03	0.40 ± 0.12
Benthic, Midwater and Surface Carnivores								
Trophic Level III								
Grouper	4	C,G,I	0.21 ± 0.01	0.66 ± 0.02	0.11 ± 0.04	0.26 ± 0.06	0.41 ± 0.05	0.27 ± 0.04
Trophic Level IV								
Ulua	4	D,G,J	0.24 ± 0.01	0.65 ± 0.02	0.19 ± 0.03	0.26 ± 0.03	0.41 ± 0.03	0.27 ± 0.03
Jacks	2	H,J	0.24 ± 0.01	0.62 ± 0.05	0.25	0.25 ± 0.02	0.38 ± 0.05	0.25 ± 0.04
Rainbow Runner	3	C,F,M	0.26 ± 0.01	0.62 ± 0.03	0.22 ± 0.02	0.32 ± 0.05	0.48 ± 0.02	0.33 ± 0.07
Snapper	5	C,F,G,H,M	0.23 ± 0.01	0.61 ± 0.05	0.11 ± 0.04	0.23 ± 0.01	0.44 ± 0.07	0.27 ± 0.03
Trophic Level V								
Mackerel	4	A,C,F,G	0.24 ± 0.01	0.54 ± 0.03	0.26 ± 0.02	0.25 ± 0.02	0.35 ± 0.02	0.26 ± 0.03
Bonito	2	F	0.29 ± 0.01	0.64 ± 0.02	0.24	0.22 ± 0.01	0.56 ± 0.01	0.32 ± 0.02
Tuna	1	G	0.24	0.63	0.25	0.24	0.40	0.21

[a] Abbreviations for atolls are A-Ailuk; C-Ailinginae; D-Bikar; S-Jemo; L-Likiep; R-Mejit; F-Rongelap; G-Rongerik; H-Taka; J-Ujelang; I-Utirik; M-Wotho.

ashed samples through modifications of published techniques, which in turn have been tested in national and international intercalibration exercises. A number of duplicate, blank, and standard samples was intermingled with the regular samples analyzed at LLNL and at the contractor laboratory. All available quality-control results for the marine samples demonstrated that the analytical performance was extremely good.

Dose Assessment Methodology

Methodology

A detailed description of the dose assessment methodology for ^{90}Sr, ^{137}Cs, $^{239,240}Pu$, and ^{241}Am is provided by Robison (1984). For the ingestion pathway, the recommended gut-to-blood transfer coefficient for plutonium isotopes is 1×10^{-4}, and for ^{241}Am it is 5×10^{-4} (ICRP 1979). Recently, however, Harrison (1983) recommended the gut-to-blood transfer coefficient be changed to 5×10^{-4} and the ICRP (1984) is presently considering this revision. The increase in dose to man by using a value of 5×10^{-4} for plutonium will therefore be considered and discussed.

Body Weights

Data from the Brookhaven National Laboratory (Greenhouse and Miltenberger, 1979 and Conard 1975) have been summarized to determine the body weight of the Marshallese people. The average adult male body weight is 72 kg for Bikini, 71 kg for Enewetak, 62 kg for Rongelap, and 70 kg for Utirik. The average, therefore, is very near the 70-kg value of reference man (ICRP 1975). As a result, we have used 70 kg as the average body weight in the dose calculations.

The average body weight for 113 adult females in the Enewetak population is 61 kg, 67 kg for 30 Utirik females, and 63 kg for 36 Rongelap females (Greenhouse and Miltenberger 1979).

Dietary Intake

The doses reported here are calculated on a daily intake of 200 g of fresh or wet weight fish muscle tissue. The 200g/d intake of fish is an upper limit from two different surveys conducted at various atolls in the Marshall Islands (Robison et. al, 1981b). The average daily intake under normal conditions would be less than 200 g/d. The dose resulting from the daily intake of anything other than 200 g can be easily calculated from the ratio of any assumed intake to the 200 g/d intake used in this report.

Percent of Whole Body Weight of Fish Tissues and Organs

The percentages of organ or tissue to whole body fresh weight were determined for several species indigenous to the atolls and are given in Table 2. As stated above, dose estimates in this paper are based only on the ingestion of concentrations associated with edible flesh. If this procedure is questioned or challenged based on new dietary information, the data in Table 2, along with the radionuclide concentrations associated with the respective tissues, can be used to construct mean concentrations in any desired assemblage of fish tissues. Two examples of the difference between flesh and reconstructed parts and whole body concentrations for fish from Bikini Atoll are shown in Table 3. Concentrations of the different radionuclides measured in the different tissues along with the values in Table 2 were used to construct the concentrations shown in Table 3. Where the difference between

TABLE 2 Percent of Whole Body Weight of Tissues and Organs for Specific Fish

Name	Muscle	Bone	Stomach contents	Viscera	Scales and skin	Liver	Gill	Reproductive Organs: Ovary	Reproductive Organs: Testes	Eyes
Acanthurus triostegus (Surgeonfish)	66.3	8.0	0.7	6.5	11.6	0.7	1.6	1.5	1.1	1.2
Crenimugil crenilabis (Mullet)	58.9	6.9	0.7	13.6	14.1	0.9	1.8	1.0	1.8	1.2
Neomyxus chaptalii (Mullet)	55.3	5.5	0.7	17.9	14.1	1.7	1.4	2.4	1.2	0.7
Mulloidichthys samoensis (Goatfish)	66.3	8.0	0.08	6.5	11.6	0.41				2.6
Aprion virescens (Snapper)	76.7	9.1	0.03	1.8	9.3	0.5	0.7	--0.23--		1.6
Mean % (all fish)	65	7.5	0.4	9.3	12.1	0.8	1.4	1.3	1.1	1.8

TABLE 3 Reconstructed Concentrations of Radionuclides in Bikini Atoll Fish.

Island	Common Name	Radionuclide	(pCi/kg wet) (see explanation below) (A)	(B)	(C)	(D)	(E)	(F)	$(\frac{A}{F})$
B-10	Convict surgeon	^{40}K	2960	2921	2610	2612	2504	2508	1.18
		$^{239,240}Pu$	0.11	1.20	2.81	3.39	27.8	28.7	3.8×10^{-3}
		^{241}Am	0.026	0.32	0.48	0.63	11.0	11.5	2.3×10^{-3}
		^{90}Sr	0.60	3.32	14.9	14.8	22.1	22.2	2.7×10^{-2}
		^{137}Cs	47.0	48.0	43.0	42.8	40.8	40.8	1.15
		^{60}Co	26.0	33.4	32.9	39.7	45.0	46.7	0.56
% reconstructed whole body weight			66.3	77.9	85.9	86.6	93.1	93.8	
B-17	Goatfish	^{40}K	3460	3281	3000	3004	3006	3004	1.15
		$^{239,240}Pu$	0.073	0.57	0.89	1.32	43.8	42.6	1.7×10^{-3}
		^{241}Am	0.030	0.20	0.41	0.55	13.8	13.8	2.2×10^{-3}
		^{90}Sr	3.2	43.1	90.0	89.7	106	105	3.0×10^{-2}
		^{137}Cs	48.0	45.0	41.2	41.2	51.9	51.9	0.92
		^{60}Co	264	296	281	308	461	473	0.56
		^{207}Bi	226	222	213	215	225	224	1.01
% reconstructed whole body weight			66.3	77.9	85.9	86.3	92.8	92.9	

(A) Muscle only concentration.

(B) Muscle + skin concentration.

(C) Muscle + skin + bone concentration.

(D) Muscle + skin + bone + liver concentration.

(E) Muscle + skin + bone + liver + viscera concentration.

(F) Muscle + skin + bone + liver + viscera + stomach content concentration = whole fish minus gills, eyes, reproductive organs.

the flesh and whole body concentration of ^{137}Cs is insignificant, there are orders of magnitude difference between the flesh and whole body concentrations of ^{90}Sr, $^{239,240}Pu$, and ^{241}Am, because these radionuclides are most concentrated in nonedible parts of the fish. Since dose estimates are proportional to concentrations, ^{90}Sr and transuranic dose rates from flesh only and whole fish ingestion will differ by orders of magnitude. It is therefore very misleading to use whole fish concentrations or even eviscerated whole fish concentrations for these radionuclides when assessing doses from fish consumption.

Results

General Comments

The concentrations of ^{90}Sr, $^{239,240}Pu$, and ^{241}Am in bone of different fish and ^{137}Cs in muscle tissue are listed in the Appendix. ^{90}Sr, $^{239,240}Pu$, and ^{241}Am were below detection limits in a large fraction of the muscle tissue samples and were detected more frequently in bone. In those instances where ^{90}Sr and $^{239,240}Pu$ were detected in both samples of muscle and bone from the pooled fish, lognormal mean wet weight ratios of muscle to bone concentrations were computed. This quotient for different fish are summarized in Table 4. Fewer ^{241}Am concentrations were available to generate these quotients, and we elected to express the mean flesh and bone concentrations relative to those of $^{239,240}Pu$ in the respective tissues. These comparisons appear in a subsequent section.

Mean bone concentrations and other computed values for ^{90}Sr and $^{239,240}Pu$ can be readily converted to muscle values using Table 4. In those instances where the muscle concentrations were below detection, the ratios in Table 4 and bone concentrations eliminate the concern on how best to handle

TABLE 4 Wet Muscle: Bone Concentration Ratios.

Fish common name	$^{239,240}Pu$	^{90}Sr
Surgeonfish	0.009	0.020
Mullet		
(*Crenimugil*)	0.017	0.010
(*Neomyxus*)	0.040	0.010
Goatfish	0.040	0.012
Grouper	<0.07	0.015
Snapper-Jack	0.058	0.015
Parrotfish	0.021	0.015
Trophic V Fish	0.038	0.015

less than or limit values when generating mean flesh concentrations at an atoll for dose estimates. All mean values, concentration factors, etc. in this text are lognormal mean values with the appropriate associated standard deviations. The results in the Appendix are sorted with respect to atoll contamination levels, trophic level, and species. One objective was to determine the degree of confidence that can be placed on the prediction of fish concentrations by the use of a concentration factor, a value relating the concentrations in any marine item to those in the supporting water in which the organism was caught, assuming a nearly uniform concentration is maintained in the environment over long periods of time.

The biological and environmental factors affecting the availability of radionuclides to fish have often been discussed (see for example Lowman et al., 1971), but simply stated, the accumulation of a radionuclide (or element) by fish may occur by direct uptake from sea water across the gills or by ingestion, from food, particulate material, and from sediment ingested with food by bottom feeding organisms. The contribution to any tissue or total body burden from each source generally cannot be determined from environmental data. However, general expressions can be written for the changes in the net accumulation of radionuclides from each pathway. These take the form of Eqs. 1-3:

$$\frac{dA_1}{dt} = a\, C_f M - \lambda_{Eff} A_1 \, , \tag{1}$$

$$\frac{dA_2}{dt} = b\, CwM' - \lambda_{Eff} A_2 \, , \tag{2}$$

$$\frac{dA_3}{dt} = cCsM'' - \lambda_{Eff} A_3 \, , \tag{3}$$

where a, b, c = are constants and represent the fractions assimilated from the source, M, M', M" = likewise assumed constant values and represent the rates of intake from the respective source,

λ_{Eff} = the excretion coefficient and is single valued regardless of the pathway, since it has been shown that radionuclides derived from sediments by fish are eliminated by a somewhat similar pattern to those taken up from seawater and food (Koyanagi et al., 1978), and

C_f, Cw, Cs = the concentrations of the radionuclide in the source terms, food, water, and sediment (or particulate material).

When equilibrium is achieved between the body burden and all pathways for a given species of fish of x years of age, dAi/dt = 0. Equation (4) is then generated by adding Eqs. (1), (2), and (3), with $A = A_1 + A_2 + A_3$; $k_1 = aM/\lambda_{Eff}$; $k_2 = bM' \lambda Eff$; $k_3 = cM''/\lambda_{Eff}$, where k_1, k_2, and k_3 are constants.

$$\frac{A}{Cw} = \frac{Cf}{Cw} \cdot k_1 + k_2 + \frac{Cs}{Cw} k_3 \qquad (4)$$

Cf:Cw can be viewed as the concentration factor for any radionuclide associated with the food and Cs:Cw as the reversible equilibrium distribution coefficient relating either or both sediment and particulate concentrations to water. Equilibrium is assumed to be rapidly established between food and water as well as sediment and water so that values for these "concentration factors" are assumed to be well established. Equation 4 can be reduced to the form of Eq. 5:

$$\frac{A}{Cw} = K = CF \quad , \qquad (5)$$

where K (= $CF_f \cdot k_1 + k_2 + Kd.k_3$) is the sum of constants and may be referred to as a field derived concentration factor (CF) from all environmentally labelled sources for any given tissue. It is easy to see from the above equations that unless the only pathway to fish is by direct accumulation from water, there should be different values for field determined concentration factors for specific radionuclides accumulated by different fish since there will be different CF_f values for species with widely different diets and fish will encounter sediment or particulate material with different sorptive properties. It is also clear that comparison of tissue concentrations should only be made to filtered seawater, since accumulation from particulate material is considered as a separate pathway. The concentration factor (CF) determined in one environment should then not be a useful value to apply in other environments. The use of generic values for fish in dose assessments should be accompanied with a large degree of uncertainty. The CF are useful site specific values, however, for establishing degrees of variability among fishes as discussed below.

Water and Sediment Concentrations

Collections of filtered seawater, surface sediments, and island soils were made along with the fish at the different atolls during 1978. Mean concentrations in the surface water and 0-4 cm surface sediments from the atoll lagoons are shown in Tables 5 and 6. In Table 6 mean atoll island surface soil concentrations are also shown. It is apparent from both the sediment and soil concentration that the atolls can be grouped according to different levels of ^{90}Sr or $^{239,240}Pu$ contamination. "A atolls" have the lowest levels of contamination and Rongelap has the highest. Radionuclide concentrations in surface sediments are lower than the current mean surface

TABLE 5 Mean 1978 lagoon filtered water concentrations

	Lagoon Surface Water (fCi/kg)						
	# of	$^{239,240}Pu$		^{241}Am		^{137}Cs	^{90}Sr
Atoll	samples	solute	particulate	solute	particulate	solute	solute
Ailuk	4	0.57 ± 0.32	0.04 ± 0.03	0.39 ± 0.19	0.05 ± 0.04	119 ± 15	91 ± 20
Ailinginae	5	0.48 ± 0.29	0.18 ± 0.11	0.28 ± 0.10	0.15 ± 0.07	131 ± 6	101 ± 15
Bikar	2	0.37 ± 0.05	0.07 ± 0.7	<.05	<.05	136 ± 19	121 ± 10
Likiep	4	0.36 ± 0.31	0.23 ± 0.30	0.17 ± 0.12	<0.11	114 ± 27	89 ± 38
Rongelap	6	1.3 ± 0.3	1.3 ± 0.3	0.54 ± 0.27	0.27 ± 0.14	171 ± 70	110 ± 7
Rongerik	4	0.45 ± 0.30	0.22 ± 0.13	0.30 ± 0.15	<.2	124 ± 14	89 ± 9
Taka	1	0.36 ± 0.17	0.06 ± 0.03	0.11 ± 0.05	0.06 ± 0.03	146 ± 24	109 ± 4
Ujelang	4	0.20 ± 0.11	0.13 ± 0.14	0.2 ± 0.1	<0.07	138 ± 29	133 ± 25
Utirik	3	0.26 ± 0.10	0.14 ± 0.07	0.2 ± 0.1	<.03	135 ± 15	109 ± 4
Wotho	4	0.42 ± 0.19	0.03 ± 0.02	0.3 ± 0.1	0.05 ± 0.01	107 ± 3	105 ± 10
Rongelap (3/81)	11	1.1 ± 0.4					
North Equatorial Pacific Surface Water 1972-78	14	0.38					
1967-82	26					140 ± 10	
Johnston Atoll (1980)	(9)					240 ± 10	

TABLE 6 Mean 1978 Near Shore Lagoon Surface Sediment (0-4 cm) and Atoll Surface (0-5 cm) Soil Concentrations

	Surface sediments					Surface soils		
	$^{239,240}Pu$		^{241}Am: $^{239,240}Pu$	^{90}Sr	^{137}Cs	$^{239,240}Pu$	^{90}Sr	^{137}Cs
Atoll	pCi/kg[a]	mCi/km^2		pCi/kg	pCi/kg	pCi/kg	pCi/kg	pCi/kg
"A Atolls"								
Ailuk	8 ± 4	0.35	0.86 ± 0.24	9 ± 4	<6	110	300	800
Likiep	8 ± 3	0.35	0.77 ± 0.32	15 ± 6	<5	43	200	700
Taka	13 ± 5	0.50	0.76 ± 0.14	12 ± 4	<5	110	700	800
Ujelang	14 ± 6	0.60	0.67 ± 0.08	17 ± 5	<5	35	200	500
Utirik	17 ± 7	0.60	0.75 ± 0.18	21 ± 7	<9	510	1400	3100
Wotho	10 ± 3	0.60	0.76 ± 0.20	16 ± 5	<6	39	200	600
"B Atolls"								
Ailinginae	56 ± 20	1.9	0.76 ± 0.09	49 ± 24	<6	2000	4900	6900
Bikar	28 ± 10	1.2	0.76 ± 0.08	29 ± 13	<7	4200	3300	300
Rongerik	82 ± 54	3.7	0.74 ± 0.08	126 ± 58	<7	1400	13100	12600
Rongelap	280 ± 150	13	0.68 ± 0.10	330 ± 360	22 ± 15	14800	55400	55700
Johnston (1980-82)					<6			

[a] All activity related to dry weight of soil and sediment.

[b] Soil values computed from Robinson et al., 1982.

soils from all atolls. Although ^{137}Cs was delivered as close-in fallout to the atolls (as shown by present higher levels in surface soil), it is absent (below gamma detection limits) from all sedimentary deposits except at Rongelap. Lagoon water concentrations of ^{137}Cs are comparable to those in the North Equatorial surface waters outside the lagoons. The ^{137}Cs delivered as local fallout to the lagoon sediments in quantities proportional to the levels now present in soils has long since mobilized from the carbonate sediments to seawater and dispersed to the open ocean. At all atolls except Rongelap, ^{137}Cs, being below detection limits in sediments, can be assumed to be in near equilibrium among the various components of the marine environment.

Lagoon water samples were collected again at Rongelap during March 1981 and analyzed for $^{239,240}Pu$. The 1981 mean concentration was within the deviation of the mean determined in 1978, which shows that these higher than oceanic background levels are real and persistent. Slow mobilization of plutonium from sediments to seawater is and has been occurring at Rongelap as it is at Bikini and Enewetak (Noshkin, 1980; Noshkin and Wong, 1980, Noshkin et al., 1984a).

Concentrations of ^{90}Sr in the surface sediments parallel those of $^{239,240}Pu$, being lowest at Ailuk, Likiep, Taka, Ujelang, Utirik, and Wotho and highest at Rongelap. Concentrations of ^{90}Sr in the lagoon water at Rongelap are not significantly different from the concentrations in seawater at the lesser contaminated atolls. Unlike $^{239,240}Pu$, any ^{90}Sr currently mobilized from sediments to seawater is masked by the throughput of global fallout concentrations associated with the North Equatorial surface waters which continuously exchange with the lagoon water mass.

Concentration ratios of ^{90}Sr, $^{239,240}Pu$, and ^{137}Cs in near shore surface sediments to those in shallow water samples are shown in Table 7. For both ^{90}Sr and $^{239,240}Pu$ the value of the ratio increases proceeding from lesser to

TABLE 7 Concentration Ratio[a]--Mean Surface Sediment to Seawater.

Atoll	^{90}Sr	^{137}Cs	$^{239,240}Pu$
Ailuk	99	<50	1.4×10^4
Likiep	168	<40	2.2×10^4
Taka	110	<30	3.6×10^4
Ujelang	128	<40	7.0×10^4
Utirik	193	<60	6.5×10^4
Wotho	155	<60	2.4×10^4
Mean "A Atolls"	1.4×10^2		4.0×10^4
Ailinginae	485	<50	1.7×10^5
Bikar	240	<50	7.5×10^4
Rongerik	1416	<50	1.8×10^5
Mean "B Atolls"	7.0×10^2		1.4×10^5
Rongelap	3.0×10^3	1.3×10^2	2.1×10^5
Johnston		<30	

[a] pCi/kg dry sediment/pCi/kg wet seawater.

more contaminated carbonate sediments. This ratio is essentially a measure of a distribution coefficient, and we expected to find the same value at all atolls, considering the similar nature of sedimentary deposits.

The radioactive contaminants present in the lagoon sediments were delivered to the region in association with complex forms of a variety of airborne particles originating from nuclear tests on coral islands of Bikini and Enewetak. Quantities of coral and other surface material were admixed with the nuclear fireball during the explosions. Some of this injected material was melted or partially melted, vaporized, or remained unchanged. During the cooling process the vaporized radionuclides could be entrained in solidified cooled matrices or condensed on surface, producing particles and agglomerates of various size, shape, and properties (as discussed in Joseph et al., 1971).

The precursors of ^{137}Cs were probably still in the gas phase when solidification was occurring, so it would be expected that the majority of ^{137}Cs was condensed on particle surfaces. Its absence today from many of the downwind sedimentary deposits then is better understood. After deposition to the sea surface most of the ^{137}Cs exchanged from the particle surfaces to seawater. There persists, however, a small fraction of ^{137}Cs more permanently fixed to the sediments, as is evident from the results of Rongelap and even more evident at Bikini and Enewetak, where concentrations are currently much higher than Rongelap in bottom sediment samples.

In laboratory experiments the range in calculated Kd values for the partitioning of ^{137}Cs from Bikini sediments to uncontaminated seawater is 10^2 - 10^4 (unpublished data). On the other hand, there is essentially no adsorption (Kd < 10) when a ^{137}Cs labelled seawater solution is shaken with uncontaminated lagoon sediments. The very weak adsorption of ^{137}Cs to carbonate sediments is best demonstrated by the results from Johnston and the "A Atolls" in Table 7, which show Kd values of less than 60, comparing bottom sediments to overlying shallow water concentrations.

There are both volatile and nonvolatile precursors in the mass 90 chain so that during the condensation and solidification process, ^{90}Sr could be found attached to surfaces or incorporated into the volume of condensing particles. $^{239,240}Pu$ and its U or Np precursors are long lived and sufficiently refractory so that most of the $^{239,240}Pu$ presently detected in sediments was distributed throughout the volume of the condensed debris that ultimately fell out over the downwind atolls. Atolls most distant from the source should have received a spectrum of particles with a smaller mean size than those delivered to close-in atolls, such as Rongelap. The smaller particles might more easily dissolve than larger particles or be reduced in size with time by erosion or other processes, thereby releasing fractions of the bound activities of ^{90}Sr and $^{239,240}Pu$ to the environments. Fractions of the mobilized activity would either remain in solution and eventually be flushed to the open ocean or adsorb on exposed fresh sediment surfaces. This explanation could account for the differences in the ^{90}Sr and $^{239,240}Pu$ concentration ratios shown in Table 7 between distant and close-in atolls. It seems from the results in Table 7 that some fraction of the radionuclides, with the possible exception of ^{137}Cs at the "A" and "B" group Atolls, is irreversibly bound in the mineral matrix of the lagoon bottom sediments (and soils) and that equilibrium conditions do not exist everywhere. This does not preclude the possibility that each lagoon may have attained steady state conditions.

TABLE 8 Mean ^{90}Sr Concentration Factors--Bone

Fish common name	A Atolls	B Atolls	A + B Atolls	Rongelap	All atolls
Trophic Level II					
Surgeonfish	2.0 ± 0.8 E02(10)[a]	4.2 ± 1.8 E02(10)	3.0 ± 2.0 E02(20)	1.3 ± 0.4 E03(7)	
Mullet	2.1 ± 0.8 E02(5)	2.5 ± 0.8 E02(4)	2.2 ± 0.9 E02(9)	3.8 ± 2.9 E03(4)	
Others	1.5 ± 0.1 E02(2)				
Trophic Level III					
Goatfish	1.9 ± 0.7 E02(8)	8.4 ± 4.6 E02(3)	3.7 ± 3.0 E02(11)	1.4 ± 0.8 E03(5)	
Other reef species	8.9 ± 0.1 E02(3)	1.8 E02(1)			
Grouper	1.4 ± 0.2 E02(2)	2.0 ± 0.1 E02(2)	1.7 ± 0.4 E02(4)		
Trophic Level IV					
Parrotfish	1.1 ± 0.3 E02(5)	1.9 ± 0.6 E02(4)	1.5 ± 0.6 E02(9)	2.2 E02(1)	1.6 ± 0.6 E02(10)
Snapper-Jack	9.0 ± 2.3 E01(4)	1.9 ± 0.1 E02(2)	1.2 ± 0.5 E02(6)	3.5 E02(1)	1.6 ± 0.9 E02(7)
Trophic Level V (Mackerel, Ulua Rainbow, Bonito, Tuna)	9.1 ± 0.3 E01(3)	1.6 ± 0.7 E02(6)	1.4 + 0.6 E02(9)	7.4 ± 2.9 E01(4)	1.2 ± 0.6 E02(13)
Representative muscle values using Table 4					
Surgeonfish	4	8	8	26	
Mullet	2	3	2	38	
Goatfish	2	10	4	17	
Parrotfish	3	3	2	3	
V fish	1	2	2	1	

[a] Value in parenthesis is the number of values averaged.

Concentrations in Fish

^{90}Sr

The mean concentration factors for ^{90}Sr in bone of different species were computed from filtered seawater concentrations and bone concentrations shown in the Appendix. Values are provided in Table 8 along with some representative muscle concentration factors computed from the bone values and the muscle-to-bone concentration ratios in Table 4. When possible, water concentrations measured nearest the fish collection site were used to compute CF values for the reef species. Mean lagoon water concentrations, shown in Table 5, were used to compute values for the pelagic species. Concentration factors are grouped for species within trophic levels from the least contaminated lagoons (group A atolls), the next level of contaminated lagoons (group B atolls), A + B lagoons, and from Rongelap Atoll, which has the highest level of ^{90}Sr associated with the near-shore sediments. When concentration factors are shown under the heading of "all atolls" differences in the computed values for fish from the A, B, and Rongelap Atolls are insignificant. Within the atoll grouping there is a slight but definite decrease in the mean bone and muscle concentration factors between the 2nd and 5th trophic levels. There do not seem to be significant differences among the bone concentration factors for species from the same trophic level.

For species belonging to the 2nd and 3rd trophic levels, there is a substantial increase in the concentration factor proceeding from lesser to more contaminated lagoons, where ^{90}Sr is more permanently fixed to the near-shore sedimentary material. Representative fish from these trophic levels are primarily bottom feeders in close contact with bottom sediments.

Inspection of the stomach contents from these fish show the presence of different quantities of ingested carbonate sedimentary material mixed with other unidentified organic material. Bone concentration factors are lowest in fish mostly removed from involvement with sediments. Some marine organisms have the ability to concentrate contaminants that are irreversibly fixed to sedimentary materials and attain higher body burdens than those attained by the same species from environments closer to equilibrium conditions.

^{137}Cs

Table 9 is arranged similarly to Table 8, showing ^{137}Cs concentration factors for fish muscle tissue. Concentrations of ^{40}K were also determined by gamma spectrometry in all fish muscle. Mean concentrations were computed for the different species and divided by 336 pCi/l (assumed ^{40}K in equatorial surface seawater) to provide reference concentration factors for comparison to the ^{137}Cs values. Concentration factors for ^{137}Cs, unlike ^{90}Sr, show more variability among species from the same trophic level, and even among species of the same family (<u>Crenimugil</u> and <u>Neomyxus</u>). Values for surgeonfish flesh are factors of 2-3 times greater than concentration factors for muscle of mullet at the A and B atolls, and the mean value for grouper is larger than that for goatfish. Concentration factors from trophic level II surgeonfish are comparable to the mean for muscle of trophic level V fish. There was no concise relationship between trophic position of the fish and their ^{137}Cs muscle concentration factor, other than noting that the concentration factors for bottom feeding fish, such as mullet and goatfish, at the A and B atolls are lower than those of pelagic species. However this is not true at Rongelap.

TABLE 9 Mean ^{137}Cs and ^{40}K Muscle Concentration Factors

Fish common name	A Atolls	B Atolls	A + B Atolls	Rongelap	All Atoll	^{40}K[a]
Trophic Level II						
Surgeonfish	1.2 ± 0.5 E02(10)	1.7 ± 0.2 E02(10)	1.5 ± 0.5 E02(20)	1.7 ± 0.7 E02(7)		10.0 (41)
Mullet						
(*Crenimugil*)	6.7 ± 1.6 E01(6)	8.1 ± 2.6 E01(3)	7.0 ± 2.0 E01(9)	1.8 ± 0.6 E02(5)		10.8 (26)
(*Neomyxus*)	3.5 ± 1.2 E01(4)	4.6 ± 0.6 E01(5)	4.2 ± 1.2 E01(9)			8.9 (19)
Rabbitfish	4.2 E01(1)					
Rudderfish	9.2 E01(1)					
Unicorn	5.9 E01(1)					
Trophic Level III						
Goatfish	5.4 ± 0.9 E01(11)	6.0 ± 0.8 E01(3)	5.6 ± 0.9 E01(14)	5.5 ± 1.2 E01(5)		11.8 (19)
Treadfin			1.1 ± 0.2 E02(3)			10.2 (7)
Flagtail	4.0 ± 1.0 E01(2)					10.2 (8)
Grouper			2.3 ± 0.9 E02(4)			12.7 (6)
Trophic Level IV						
Parrotfish			1.3 ± 0.3 E02(7)	1.3 E02(1)		11.7 (14)
Jack	1.9 ± 0.8 E02(2)					11.8 (9)
Snapper			1.4 ± 0.3 E02(5)			12.5 (8)
Trophic Level V (Ulua-Rainbow-Mackerel, Tuna, Bonita)					1.6 ± 0.4 E02(14)	11-14

[a] Seawater concentration assumed -336 pCi/l. Number of samples averaged in parenthesis. (^{40}K mean value determined by averaging some values in fish from Enewetak-Bikini).

At Rongelap the ^{137}Cs CF computed for mullet muscle is comparable to that for surgeonfish and exceeds the mean value computed for all pelagic species. As with ^{90}Sr, higher concentration factors are found associated with fish at atolls where a fraction of the radionuclide is still irreversibly fixed to sedimentary material. Body burdens attained by bottom feeding fish are higher than those in the same species from environments closer to equilibrium conditions.

The concentration factors determined for the different species at the A atolls were used to predict concentrations in the flesh of fish from Johnston Atoll collected in 1980 and 1982. The average water concentrations at Johnston Atoll during these periods were 0.24 and 0.17 pCi/l, respectively. These concentrations are higher than any encountered in the Marshall lagoon waters during 1978. Predicted concentrations are shown along with measured flesh concentrations in Table 10. There is excellent agreement between predicted and measured values for the different species representing three trophic levels. However, had the same values been applied to compute concentrations in the flesh of bottom feeding fish at Rongelap, Bikini, or Enewetak, much lower concentrations would have been predicted than were measured. Examples are shown in Table 11 where the predicted concentrations in muscle of mullet from Rongelap, Bikini, and Enewetak are compared to measured concentrations using the concentration factor determined at the "A" atolls.

The ^{137}Cs flesh concentration factors for bottom feeding fish from the A and B atolls can be used with some degree of accuracy to predict concentrations in flesh from water concentrations at those atolls in near equilibrium conditions with respect to partitioning of ^{137}Cs among environmental components. These concentration factors for ^{137}Cs (as well as for ^{90}Sr) cannot, however, be used with any degree of accuracy for bottom feeding fish

TABLE 10 ^{137}Cs Concentrations Measured and Predicted in Fish Muscle From Johnston Atoll

				pCi/kg wet	
Station	Mo/Yr	Common name	CF[c]	Measured concentration flesh	Predicted concentration flesh
3	3/1980[a]	Surgeonfish	120 ± 50	30 ± 1	29 ± 5
6	3/1980	Surgeonfish		30 ± 1	29 ± 5
8	3/1980	Surgeonfish		33 ± 2	29 ± 5
3	3/1980	Mullet *Neomyxus*	35 ± 12	6.8 ± 0.4	8 ± 3
8	3/1980	Mullet *Neomyxus*		7.4 ± 1.4	8 ± 3
10	3/1980	Mullet *Neomyxus*		6.8 ± 0.6	8 ± 3
Sand Is	3/1980	Goatfish	54 ± 9	11 ± 2	13 ± 2
Sand Is	3/1980	Jack	190 ± 80	22 ± 1	46 ± 19
3	6/1982[b]	Surgeonfish	120 ± 50	19 ± 3	20 ± 8
6	6/1982	Surgeonfish		13 ± 1	20 ± 8

[a] Mean water concentration during collection--0.24 ± 0.02 pCi/l.

[b] Mean water concentration during collection--0.17 ± 0.02 pCi/l.

[c] Value developed at "A" atolls in the Marshall Islands (see Table 9).

TABLE 11 Predicted ^{137}Cs Muscle Concentrations in Mullet Using the Value of the ^{137}Cs Concentration Factor Developed for this Species at the A Atolls

Atoll collection date	Water pCi/l	Predicted muscle from CF (pCi/kg wet)	Measured (pCi/kg wet)	Fraction predicted
[a] B-1 1/77	0.35	23	263	0.09
B-1 11/78	0.39	26	397	0.07
B-2 1/77	0.35	23	379	0.06
B-6 2/81	0.25	17	60	0.28
B-13 1/77	0.19	12	21	0.57
B-17 11/78	0.51	34	88	0.38
E-2 4/76	0.34	22	211	0.10
E-10 3/78	0.23	15	211	0.07
E-33 4/76	0.17	11	14	0.79
E-24n 11/78	0.44	29	262	0.11
F-9 11/78	0.13	9	16	0.56
F-23 11/78	0.17	12	21	0.57
F-1 11/78	0.13	8	27	0.29

[a] B = Bikini Atoll; E = Enewetak Atoll; F = Rongelap Atoll.

from environments where fractions of the radionuclides are still found irreversibly bound to sediments. Clearly the bottom fish from these environments demonstrate a capability of deriving fractions of their body burden from these labelled sediments. The concentrations associated with the sediments are not in equilibrium with those in the water. Although the ambient concentrations in water, sediment, and food are not greatly fluctuating (approximating steady state conditions), a generic CF for any species of fish, determined in environments close to equilibrium conditions, would grossly underestimate measured concentrations at Rongelap, Bikini, and Enewetak when referenced to water concentrations.

^{239,240}Pu and ^{241}Am

Bone concentration factors were computed from the available data for ^{239,240}Pu and ^{241}Am and are shown in Tables 12 and 13. The errors associated with the mean values for some species are somewhat greater than those for ^{90}Sr and ^{137}Cs; however, the trends among the mean values are of interest. A large difference in the plutonium and americium bone values is seen for trophic level IV species; parrotfish feed on live coral heads, while the snapper and jacks are mid-to-surface water carnivores. Live coral has been shown to accumulate plutonium from the environment with concentration factors exceeding 10^3 (Noshkin et al., 1975). The differences between the bone CF values for these species within the same trophic level must be related to feeding habits. With the exception of the parrotfish values, there is a decrease in the CF values for bone and muscle between trophic II and trophic V fish. As noted for ^{90}Sr there is an increase in the value of the bone CF for ^{239,240}Pu and ^{241}Am,

TABLE 12 Mean ^{239,240}Pu bone concentration factors.

Fish common name	A atolls	B atolls	A + B atolls	Rongelap	All atolls
Trophic Level II					
Surgeonfish	1.1 $\pm$ 1.0 E04(8)	2.9 $\pm$ 1.9 E04(9)	2.1 $\pm$ 2.4 E04(17)	3.4 $\pm$ 2.4 E04(7)	-- --
Mullet	7.9 $\pm$ 2.8 E03(12)	1.3 $\pm$ 1.4 E04(7)	9.4 $\pm$ 6.4 E03(19)	2.8 $\pm$ 1.2 E04(6)	-- --
Others	1.3 $\pm$ 0.6 E03(3)	-- --	-- --	-- --	-- --
Trophic Level III fish	3.0 $\pm$ 2.1 E02(7)	4.6 $\pm$ 6.9 E02(3)	3.4 $\pm$ 3.0 E02(10)	4.1 $\pm$ 2.8 E02(5)	3.2 $\pm$ 2.5 E02(15)
Goatfish	2.9 $\pm$ 1.8 E02(4)	-- --	-- --	4.1 $\pm$ 2.8 E02(5)	-- --
Trophic Level IV					
Parrotfish	-- --	-- --	5.2 $\pm$ 4.3 E03(8)	1.2 E04(1)	-- --
Snapper-Jack	-- --	-- --	-- --	-- --	2.7 $\pm$ 1.1 E02(5)
Trophic Level V	-- --	-- --	-- --	-- --	2.0 $\pm$ 2.2 E02(9)
Representative Muscle values using Table 4					
Surgeonfish	100	260	190	300	--
Mullet	240	260	--	840	--
Goatfish	12	--	--	16	--
Parrotfish	--	--	110		
Snapper-jack	--	--	--	--	15
Trophic V fish	--	--	--	--	7

TABLE 13 Mean ^{241}Am bone concentration factors.

Fish common name	A atolls	B atolls	A + B atolls	Rongelap	All atolls
Trophic Level II					
Surgeonfish	1.9 ± 2.3 E03(7)	1.8 ± 1.5 E03(7)	1.9 ± 1.9 E03(14)	6.7 ± 5.9 E03(6)	-- --
Mullet	1.0 ± 0.9 E03(5)	4.2 ± 3.1 E03(3)	2.2 ± 3.0 E03(8)	3.7 ± 5.0 E03(4)	-- --
Trophic level III	-- --	-- --	-- --	-- --	5.4 ± 4.2 E02(11)
Trophic level IV	-- --	-- --	1.7 ± 1.7 E03(10)	1.8 E03(1)	1.7 ± 1.7 E03
Parrotfish	-- --	-- --	2.9 ± 1.1 E03(6)	1.8 E03(1)	-- --
Snapper-Jack	-- --	-- --	6.6 ± 2.1 E02(4)	-- --	-- --
Trophic level V	-- --	-- --	-- --	-- --	5.4 ± 3.6 E02(5)

most noticeable in trophic II fish, proceeding from lesser to more contaminated lagoons. This is the only general feature common to all the man-made radionuclides determined.

In Table 14, ^{241}Am muscle and bone concentrations and concentration factors are compared to those of $^{239,240}Pu$. Insufficient muscle data is available to generate a quotient for trophic V fish, but the values show a trend in the abilities of certain fish to accumulate plutonium and americium from the environment. ^{241}Am is enriched in bone over $^{239,240}Pu$ in higher trophic level fish, since there is a general increase in the concentration quotient proceeding from trophic II to trophic V fish. These are the only observations showing these trends in different species of fish; further study is required on how these relative concentrations are attained.

Dose Assessment

Average muscle radionuclide concentrations, shown in Table 15, were determined for the various fish from each group of atolls. In those instances where muscle concentrations of ^{90}Sr, $^{239,240}Pu$, or ^{241}Am were below detection, muscle concentrations were calculated from bone using the mean ratios described in the text. Since dietary studies showed no preference for any specific species, the average radionuclide concentration for all species of fish flesh was used with the average daily intake, the average biological residence times, and the fractional depositions to calculate the maximum annual dose rates and 30-y integral doses. Mean fish concentrations were multiplied by the average intake of 200 g/d of fish to obtain pCi/d ingested in Table 16.

The maximum annual mean dose rate for the whole body is defined as the dose rate in that year when the ingestion dose from ^{137}Cs is a maximum, and for

TABLE 14 Mean $^{241}Am:^{239,240}Pu$ Concentrations in Bone and Muscle and Comparative Bone Concentration Factors

Fish common name	$^{241}Am:^{239,240}Pu$ Muscle ratio	$^{241}Am:^{239,240}Pu$ Bone ratio	Bone Mean concentration factors Pu	Am	Atolls
Trophic level II					
Surgeonfish	0.19	0.06 ± 0.04	2.1 E04	1.9E03	A + B atolls
Mullet	0.13	0.10 ± 0.08	9.4 E03	2.2E03	A + B atolls
Trophic level III	0.45	0.41 ± 0.27	3.2 E02	5.4E02	All atolls
Trophic level IV					
Parrotfish	0.48	0.52 ± 0.40	5.2 E03	2.9E03	A + B atolls
Snapper-jack		1.2 ± 0.8	2.7 E02	6.6E02	All atolls
Trophic level V	--	2.0	2.0 E02	5.4E02	All atolls

TABLE 15 Mean and Range of Fish Flesh Concentrations at the Grouped Atolls

Atolls	pCi/kg wet ^{137}Cs	^{90}Sr	$^{239,240}Pu$	^{241}Am
Group A Atolls				
Mean	15	0.32	0.065	0.011
Range	4-38	0.05-0.9	0.005-0.19	
Group B Atolls				
Mean	18	0.60	0.092	0.011
Range	5-43	0.05-1.0	0.005-0.3	
Rongelap				
Mean	23	1.0	0.44	0.032
Range	6-61	0.06-2.8	0.008-1.4	

TABLE 16 Calculated Ingestion Rate in 1978 and Maximum Annual Mean Dose Rate of ^{90}Sr, ^{137}Cs, $^{239,240}Pu$ and ^{241}Am in the Northern Marshall Islands.

Atolls	pCi/d based on 200 g/d intake of fish				Maximum annual dose rate (mrem/y)	
	^{137}Cs	^{90}Sr	$^{239,240}Pu$	^{241}Am	Whole body	Bone marrow
A atolls	3	0.064	0.013	0.002	0.059	0.067
B atolls	3.6	0.12	0.018	0.003	0.071	0.084
Rongelap	4.6	0.2	0.088	0.006	0.091	0.184

bone marrow, when the ingestion dose from the sum of all radionuclides is a maximum. Because of the dose buildup from ^{90}Sr, $^{239,240}Pu$, and ^{241}Am and the continuously decreasing dose after the first year for ^{137}Cs, the bone-marrow maximum annual dose rate can occur in a different year than the whole-body maximum annual dose rate. The radionuclide doses are calculated using the measured radionuclide concentrations and assuming that they change with time only by physical decay. The maximum annual dose rates for the A and B atolls and Rongelap are listed in Table 16. The whole-body dose rates are fractions of a mrem/y and range from 0.06 mrem/y at the A atolls to 0.09 mrem/y at Rongelap. The bone-marrow dose rates range from 0.07 to 0.18 mrem/y. For perspective, these maximum annual dose rates can be compared to the current Federal Guidelines for whole body and bone marrow of 500 mrem/y for an individual and 170 mrem/y for a population.

The dose commitment or 30-y integral doses and the contribution of each radionuclide to the 30-y integral dose from fish consumption are shown in Table 17 along with the difference caused by increasing the value of the gut transfer coefficient for plutonium to 5×10^{-4}. The 30 y integral doses range from 0.03 to 0.08% of the Federal 30-y guideline of 5 rem. ^{137}Cs is the largest contributor to the small doses from the marine food chain. When the gut transfer coefficient of 5×10^{-4} is used for plutonium, the fractional contribution from the transuranic is 30% of the total dose at Rongelap and less than 15% at the other atolls. There is little reason to single out the transuranics as potential hazards in the fish ingestion pathway at these atolls, unless the normally nonedible parts of fish become part of the diet. Since ^{137}Cs concentrations are notably less in bottom-feeding fish, such as mullet and goatfish, these species would be recommended to comprise the major fraction of marine diets in order to minimize the accumulation of ^{137}Cs from the fish ingestion pathway.

TABLE 17 The 30 yr integral dose in mrem for each radionuclide for an intake of 200 g/d of fish flesh.

Atolls	^{90}Sr Bone marrow	^{137}Cs Whole body	^{137}Cs Bone marrow	$^{239,240}Pu$ Bone marrow A[a]	$^{239,240}Pu$ Bone marrow B[a]	^{241}Am Bone marrow
A atolls	0.20	1.4	1.4	0.033	0.16	0.025
B atolls	0.37	1.6	1.6	0.045	0.23	0.025
Rongelap	0.62	2.1	2.1	0.27	1.1	0.0077

	Total whole body	Total bone marrow	Transuranics % of bone dose
A atolls	1.4	1.7 (1.3)[b]	3.4 (10)[b]
B atolls	1.6	2.0 (2.2)	3.5 (12)
Rongelap	2.1	3.0 (3.9)	9.9 (30)

[a] A column assumes gut transfer coefficient of 1×10^{-4} for Pu.
B column assumes gut transfer coefficient of 5×10^{-4} for Pu.

[b] Values in parenthesis are total dose and % of bone dose with 5×10^{-4} as the gut transfer coefficient for Pu.

Discussion

It is clear that bottom-feeding fish have the ability to extract radionuclides that are irreversibly bound to sedimentary deposits and attain tissue burdens of ^{90}Sr, $^{239,240}Pu$, and ^{137}Cs, which are larger than the concentrations in the same species from environments where near equilibrium conditions have been established. In studies with parrotfish at Enewetak Atoll, Smith and Paulson (1974) note that during feeding the entire interior of the gut of the fish was more acidic than during periods of nonfeeding. These authors suggest the possibility that some of the calcium carbonate particles ingested by the fish could dissolve in the gut, and they show that particles removed from the large intestines were reduced rapidly in size when placed in buffered solutions having the same acidic pH as the intestine. Smith and Paulson (1975) also report that the intestinal mucosa of surgeonfish and parrotfish from Enewetak contained an order of magnitude higher carbonic anhydrase activity than predators that take in very little calcium carbonate while feeding. There was no demonstrable enzyme activity in the sea water, algae, or coral particles normally ingested by the fish; therefore, the carbonic anhydrase, which affects the breakdown of H_2CO_3 formed from the solution of $CaCO_3$ in the presence of weak stomach acids, in bile, mucosa, and gut contents, is all endogenous. The authors state that their results are consistent with the hypothesis that calcium carbonate ingestion results in solution of calcium carbonate within the gut of these fish. During September 1984, pH measurements were made in slurries of stomach and intestinal contents from freshly caught mullet, surgeonfish, goatfish and selected carnivores. The gut content pH ranged from 5.0-2.7 in mullet; 7.0-7.6 in surgeonfish; 6.6-7.7 in goatfish and 5.2-6.8 in three species of lagoon carnivores. We could not demonstrate that the contents from feeding surgeonfish were sufficiently acidic to dissolve quantities of $CaCO_3$ but the contents from the other species of fish were sufficiently acidic during feeding to breakdown quantities of ingested $CaCo_3$. We reason that the above describes the only mechanism to adequately explain the higher tissue burdens in bottom feeding fish at atolls where radionuclides are more strongly bound to the calcium carbonate sedimentary

particles. Fractions of ^{137}Cs, $^{239,240}Pu$, ^{241}Am, and ^{90}Sr inversibly bound to the sediments (most noteable for the transuranics and ^{90}Sr at Rongelap) may be released by the acidic digestive juices in the gut of bottom-feeding and coral-feeding fish. The radionuclides ultimately pass across the gut wall with any other nutrients or pollutants dissolved from the calcium carbonate matrix during digestion. The radionuclides released in the gut during digestion are not in equilibrium with the surrounding sea water. This would account for the higher than near "equilibrium" concentration factors when tissue burdens of sediment ingesting fish are compared to water concentrations.

For some samples of mullet, sufficient material was extracted from the gut to provide reliable concentrations of ^{90}Sr and ^{239}Pu associated with the gut contents. Assuming that there is an equilibrium CF_1 related to water and another CF_2 related to the quantities bound to the ingested material, and that the concentration factors are independent of the concentrations in each source, the tissue concentration can be expressed with a two-source model equation:

$$C = CF_1 . W + CF_2 . I \; , \qquad (6)$$

where C is any tissue concentration and W and I are the water and ingested concentrations. The assumption must also be made that the concentrations associated with the gut material when the fishes were caught are representative of the levels normally encountered over their lifetime.

The two concentration factors CF_1 and CF_2 can be estimated from a regression of C on W and I. Concentration data for ^{90}Sr in 10 samples of mullet bone, stomach contents, and lagoon water are shown in Table 18. The regression analysis yields a value of 124 for CF_1 and 0.16 for CF_2 with a correlation coefficient of 0.987. The percent of ^{90}Sr in bone derived from the "equilibrium" environment and the ingested source of irreversibly bound ^{90}Sr are computed and shown in Table 18. The contribution of ^{90}Sr in bone derived from the sedimentary source is variable and ranges from 14% to 93% of the total concentration measured in bone of mullet at the different atolls. The mean total percent derived from the "irreversible sedimentary phase" increases between the lesser contaminated atolls and Rongelap. The "equilibrium" bone CF for mullet is 124, a factor of 2 lower than the average value determined for mullet at the A atolls. Even at these atolls there remains a fraction of the ^{90}Sr more firmly fixed within the matrix of the original fallout particles. The "equilibrium" muscle CF for ^{90}Sr in mullet has a value of 1, which is probably the best representative mean value for muscle of all species. Insufficient stomach content data were available for ^{137}Cs to attempt any meaningful correlation of the previous type. However, at Bikini and Enewetak the two-source model of the type used for ^{90}Sr can be used to explain much of the variation in the ^{137}Cs fish concentration data at these atolls (Noshkin et al., 1984b).

Data for $^{239,240}Pu$ shown in Table 19 were also used with the two-source model to generate values for CF_1 and CF_2. The regression equation resulted in values of 5650 and 0.053 for CF_1 and CF_2, respectively. The variance however was much larger for $^{239,240}Pu$ than for ^{90}Sr. Percentages of $^{239,240}Pu$ in bone derived from the two sources at the A, B, and Rongelap Atolls are shown in the last columns of Table 19. The percent in bone derived from the "equilibrium" labelled environment decreases from the lesser contaminated atolls to Rongelap,

TABLE 18 Concentrations of ^{90}Sr in Bone and Stomach Contents of Mullet and Seawater.

Estimated Amounts in Bone from the Two Source Model

	^{90}Sr (pCi/kg wet)			Concentrations in bone from source 1 & 2 using two-source model			% of bone concentration derived from source 1 & 2	
Island	Bone	Stomach contents	Water	Source[a] 1	Source[b] 2	Source 1 & 2	Source[a] 1	Source[b] 2
A Atolls								
A53	23	61	0.091	11.2	9.6	21	54	46
L37	13	40	0.089	11.0	6.4	17	62	38
H1	40	37	0.109	13.5	5.9	19	70	30
H4	19	37	0.109	13.5	5.9	19	70	30
M12	11	14	0.104	12.9	2.2	15	86	14
			Mean A Atolls				68 $\pm$ 12	32 $\pm$ 12
B Atolls								
C27	27	123	0.101	12.5	19.6	32	39	61
C27	14	67	0.101	12.5	10.6	23	54	46
D4	29	127	0.121	15	20.2	35	42	58
G11	30	78	0.089	11	12.4	23	46	54
			Mean B Atolls				45 $\pm$ 7	55 $\pm$ 7
Rongelap								
F23	212	1234	0.118	14.6	196	211	7	93

[a] Contribution from the equilibrium labelled environment.

[b] Contribution from the irreversibly bound ^{90}Sr associated with material ingested.

TABLE 19 Concentration of $^{239,240}Pu$ in Bone and Stomach Contents of Mullet and Seawater.

Estimated Amounts in Bone of Mullet from the Two Source Model.

Island	$^{239,240}Pu$ (pCi/kg wet)			Concentration in bone from source 1 & 2 using 2 source model			% of bone concentration derived from source 1 & 2	
	Bone	Stomach contents	Seawater x 10^3	Source[a] 1	Source[b] 2	Source 1 & 2	Source[a] 1	Source[b] 2
A Atolls								
A1	3.3	10.9	0.57	3.2	0.6	3.8	84	16
A53	5.7	17.4	0.57	3.2	0.9	4.1	78	22
L37	1.4	7.1	0.36	2.0	0.4	2.4	83	17
L55	1.9	8.9	0.36	2.0	0.5	2.5	80	20
H1	4.5	18	0.36	2.0	1.0	3.0	67	33
H4	2.8	18	0.36	2.0	1.0	3.0	66	34
H5	2.8	10	0.36	2.0	0.5	2.5	80	20
				Mean A Atolls			77 ± 7	23 ± 7
B Atolls								
C27	17	56	0.48	2.7	3.0	5.7	47	53
C27	4.9	38	0.48	2.7	2.0	4.7	57	43
D1	1.3	27	0.37	2.1	1.4	3.5	60	40
D1	1.1	76	0.37	2.1	4.0	6.1	34	66
D4	2.0	38	0.37	2.1	2.0	4.1	51	49
G11	2.5	23	0.45	2.5	1.2	3.7	32	68
				Mean B Atolls			47 ± 1	53 ± 11
Rongelap								
F23	43	427	1.24	7.0	23	30	24	76
F47	16	364	1.14	8	19	27	29	71
				Mean Rongelap			26 ± 2	74 ± 2

[a] Contribution from the equilibrium labelled environment.
[b] Contribution from the irreversibly bound ^{90}Sr associated with material ingested.

with a corresponding increase in the fraction of bone $^{239,240}Pu$ derived from the sources associated with the sediments. Percentages derived from each source at the different atolls are close in value to those of ^{90}Sr shown in Table 18. The two-source model shows that both pathways contribute to the internal body burdens of bottom-feeding fishes; but, when the levels of contamination increase in bottom sediments, the pathway involving solution of components within the calcium carbonate matrix in the gut of fish dominates. This identified pathway clearly deserves further study to understand the accumulation by bottom-feeding fish of any pollutant, stable element, or nutrient incorporated into calcuim carbonate deposits at coral atolls. Pollutants irreversibly fixed to carbonate sediments are not isolated from biological cycles in the ocean. Higher trophic level species that do not rely on sediments or coral for this source of food show no such increasing trend in the values for the concentration factors between differently contaminated atolls. Concentration factors computed by relating tissue concentrations for these species to water concentrations can be used in generic model applications.

Summary

Concentrations of ^{90}Sr, ^{137}Cs, $^{239,240}Pu$, and ^{241}Am were determined in tissues of fish, near-shore surface sediments, and seawater from several atolls contaminated with close-in fallout debris generated at the Pacific Proving Grounds in the late 1940s through the late 1950s. Sediment-to-lagoon water concentration ratios for these radionuclides (a measure of the sediment/water distribution coefficient) increased proceeding from the lesser contaminated atolls to Rongelap. Fractions of the radionuclides still detected in the marine environment are irreversibly bound with the mineral matrix of the sediments, and equilibrium conditions do not exist at all atolls.

The atolls were sorted with respect to present contamination levels detected in the surface sediments. Mean radionuclide CFs were computed for bone and muscle of the fish representing trophic levels II-V from the groups of atolls by relating the measured tissue concentrations to those in filtered lagoon seawater. Values of ^{90}Sr concentration factors for bone and muscle of fish decrease between the 2nd and 5th trophic level. The concentration factor for bone of bottom feeding fish is largest at atolls where ^{90}Sr is more permanently fixed to the near shore sedimentary material. There was no unique relationship between trophic levels of the fish and the ^{137}Cs muscle concentration factor. Some of the variability in the ^{137}Cs concentration factors is best related to differences in diet, with concentration factors being lower for bottom feeding fish than for pelagic species. Higher concentration factors are also found for ^{137}Cs at atolls where the radionuclide is still detected in bottom sediments.

Values of $^{239,240}Pu$ and ^{241}Am concentration factors for bone followed the same trend noted for ^{90}Sr, decreasing between II and V trophic level species. An exception to this was noted for the coral grazing parrotfish (trophic level IV). The transuranic concentration factors for bone were nearly an order of magnitude larger than the value of the CF for other trophic level IV species. Ratios of ^{241}Am to $^{239,240}Pu$ increased in bone and muscle between 2nd and 5th trophic level species. Both $^{239,240}Pu$ and ^{241}Am concentration factors are greater for bottom-feeding species from the more contaminated atolls than found for the same species at the lesser contaminated atolls. This feature is common for all the long-lived man-made radionuclides determined in this study. A partial ranking of the radionuclide concentration factors in muscle for some species is provided below.

Surgeonfish muscle concentration factors

A atolls $^{137}Cs \geq {}^{239,240}Pu > {}^{241}Am > {}^{90}Sr$

B atolls $^{137}Cs \geq {}^{239,240}Pu > {}^{241}Am > {}^{90}Sr$

Rongelap $^{239,240}Pu > {}^{137}Cs > {}^{241}Am > {}^{90}Sr$

Mullet muscle concentration factors

A atolls $^{239,240}Pu > {}^{137}Cs > {}^{241}Am > {}^{90}Sr$

Goatfish muscle concentration

A atolls $^{137}Cs \geq {}^{239,240}Pu > {}^{241}Am > {}^{90}Sr$

Trophic V species

All atolls $^{137}Cs > {}^{241}Am > {}^{239,240}Pu > {}^{90}Sr$

This ranking shows there is no precise ordering of the values of concentration factors for all fish, which negates the use of a single generic value for all fish species. The ordering is altered by species, trophic level, and degree of bottom sediment contaminations.

Bottom-feeding fish have the ability to extract radionuclides that are irreversibly bound to sedimentary deposits and attain tissue burdens that are larger than the concentrations found in the same species from environments where near-equilibrium conditions have been established. The values for the concentration factors generated at the lesser contaminated atolls cannot be used with water concentrations to generate reliable estimates of concentrations in tissues of species from the more contaminated lagoons. An explanation for this anomaly is that some bottom- or coral-feeding fish with diets containing, in part, carbonate material have the ability to lower their gut pH during feeding, which results in dissolution, within the gut, of a fraction of the calcium carbonate ingested with food. Fractions of the ^{137}Cs, ^{90}Sr, $^{239,240}Pu$, and ^{241}Am presently fixed within the calcium carbonate matrix are released by the digestive juices and can pass across the gut wall. A two-source model is used to compute the contribution of ^{90}Sr and $^{239,240}Pu$ from the equilibrium labelled environment and the sedimentary "bound" source term.

The percent of the body burden for mullet from the sediment bound source increases between the lesser to more contaminated atolls. At Rongelap it is the most dominate source term contributing to radionuclide body burden in bottom-feeding fish. As the quantities of irreversibly bound radionuclides are reduced with time through radioactive decay or by other biogeochemical processes, the concentration factors now determined at the more contaminanted atolls will decrease and eventually be reduced to the values now seen at the lesser contaminated atolls. Concentration factors for higher trophic level species, which do not rely on sediments or coral for the source of food, show no trend or change in value between differently contaminated atolls. Concentration factors computed by relating tissue concentrations for these species to water concentrations can be used in generic model applications.

Computed dose rates and 30-y integral dose from fish consumption are very low; they are small fractions of the current Federal Guidelines. ^{137}Cs is presently the largest contributor to the small dose from the marine fish consumption pathway, with the transuranics contributing from 2%-30% of the total dose. The range in the percentage is due to the atoll considered and the value used for the $^{239,240}Pu$ gut transfer coefficient. There is little reason to single out the transuranics as potential hazards in the marine fish ingestion pathway at these atolls unless fish parts not normally eaten become part of man's diet.

There were a number of still unanswered questions developed from this study that might be worthy of future research. For example, there is a need to verify the release and rate of release for sediment bound radionuclides to solution under conditions of different pH measured in the guts of fish. Comparison of fish tissue concentrations to sediments rather than water may provide more reasonable correlation between environmental concentrations and fish body burdens. More needs to be known about the metabolism of plutonium and americium and the reasons for the observed differences in tissue concentrations for fish of different trophic levels.

Acknowledgments

We thank Reynold De Brum, Trust Territories of the Pacific and presently employed by DOE, for his aid in the biological collections made in 1978. We also acknowledge the assistance during phases of the program from R. Spies, P. Davis, L. Gagley, D. Hosmer, J. McNabb, G. Holladay, T. Jokela, J. L. Brunk and K. V. Marsh, Lawrence Livermore National Laboratory. This work was performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract W-7405-ENG-48.

REFERENCES

Conard, R. A., 1975, A Twenty Year Review of Medical Findings in a Marshallese Population Accidentally Exposed to Radioactive Fallout, BNL-50424, Brookhaven National Laboratory, Upton, NY.

Greenhouse, N., and R. Mitlenberger, 1979, Brookhaven National Laboratory, Upton, NY cited in Lawrence Livermore National Laboratory, UCRL-52853 (Pt. 3).

Harrison, J. D., 1983, The Gastrointestinal Absorption of Plutonium, Americium and Curium, Rad. Proc. Dosimetry, 5: 19-35.

Hiatt, R. W., and D. W. Strasburg, 1965, Ecological Relationships of the Fish Fauna on Coral Reefs of the Marshall Islands, Ecol-Monographics 30, 65 pp.

International Commission on Radiological Protection, 1975, Report of the Task Group on Reference Man, ICRP Publ. 23, Pergamon Press, Oxford.

International Commission on Radiological Protection, 1979, Annals of the ICRP, Limits for Intakes of Radionuclides by Workers ICRP Publ. 30, Pergamon Press, Oxford, pt. 1.

International Commission on Radiological Protection, 1984, Annals of the ICRP, Statement from the 1983 Washington Meeting of the ICRP, ICRP Publ. 39, Pergamon Press, Oxford, vol. 14, no. 1, pp. 1-7.

Joseph, A. B., P. F. Gustafson, I. R. Russel, E. A. Schuert, M. L. Volchok, and A. Tamplin, 1971, Sources of Radioactivity and their Characteristics, in Radioactivity in the Marine Environment, National Academy of Sciences, Washington, D.C., chpt. 2, pp. 6-41.

Koyanagi, T., M. Nakakara, and M. Iimura, 1978, Absorption of Sediment-Bound Radionuclides through the Digestive Tract of Marine Demersal Fishes, J. Radiat. Res., 19: 295-305.

Lowman, F. G., T. R. Rice, and F. A. Richards, 1971, Accumulation and Redistribution of Radionuclides by Marine Organisms, in Radioactivity in the Marine Environment, National Academy of Sciences, Washington, D.C., chpt. 7, pp. 161-199.

Noshkin, V. E., K. M. Wong, R. J. Eagle, and C. Gatrousis, 1975, Transuranics and Other Radionuclide in Bikini Lagoon: Concentration Data Retrieved from Aged Coral Sections, Lunnol. & Oceans, 20: 729-742.

Noshkin, V. E., 1980, Transuranium Radionuclides in Components of the Benthic Environment of Enewetak Atoll, in Transuranic Elements in the Environment, W. C. Hanson (Ed.), DOE/TIC-22800, U.S. Department of Energy, Washington, D.C., pp. 578-601.

Noshkin, V. E., and K. M. Wong, 1980, Plutonium Mobilization from Sedimentary Sources to Solution in the Marine Environment, in Marine Radioecology, Proceedings of 3rd Nuclear Agency Seminar, Tokyo, Oct. 1-5, 1979, Nuclear Energy Agency/OECD, Paris, pp. 165-178.

Noshkin, V. E., R. J. Eagle, K. M. Wong, and T. A. Jokela, 1981, Transuranic Concentrations in Reef and Pelagic Fish from the Marshall Islands, in Impacts of Radionuclide Releases into the Marine Environment, Symposium Proceedings, Vienna, Oct. 1980, International Atomic Energy Agency, Vienna, pp. 293-317.

Noshkin, V. E., K. M. Wong, T. A. Jokela, J. L. Brunk, and R. J. Eagle, 1984a, Plutonium and Americium Behavior in Coral Atoll Environments, in Wiley-Interscience Series Wastes in the Ocean, 4th International Ocean Disposal Symposium, Plymouth, England, April 1983, in press.

Noshkin, V. E., K. M. Wong, R. J. Eagle, T. A. Jokela, and J. L. Brunk, 1984b, Gamma Emitting Radionuclides in Species of Fish from Enewetak and Bikini, Lawrence Livermore National Laboratory, Livermore, CA, in preparation.

Robison, W. L., V. E. Noshkin, W. A. Phillips, and R. J. Eagle, 1981a, The Northern Marshall Islands Radiological Survey: Radionuclide Concentrations in Fish and Clams and Estimated Doses via the Marine Pathway, UCRL-52853 (Pt. 3), Lawrence Livermore National Laboratory, Livermore, CA.

Robison, W. L., W. A. Phillips, M. E. Mount, B. R. Clegg, and C. L. Conrado, 1981b, Reassessment of the Potential Radiological Doses of Residents Resettling Enewetak Atoll, UCRL-53066, Lawrence Livermore National Laboratory, Livermore, CA.

Robison, W. L., M. E. Mount, W. A. Phillips, C. L. Conrado, M. L. Stuart, and C. E. Stoker, 1982, The Northern Marshall Islands Radiological Survey: Terrestrial Food Chain and Total Dose, UCRL-52853, (Pt. 4), Lawrence Livermore National Laboratory, Livermore, CA.

Robison, W. L., 1984, Radiological Dose Assessments of Atolls in the Northern Marshall Islands, in Proceedings of the 19th Annual Meeting of the National Council on Radiation Protection and Measurements, NAS, Washington, D.C., April 1983, NCRP Proc. 5, pp. 40-82.

Smith, R. L., and A. C. Paulson, 1974, Food Transit Times and Gut pH in Two Pacific Parrotfish, Copeia, 3: 796-799.

Smith, R. L., and A. C. Paulson, 1975, Carbonic Anhydrase in some Coral Reef Fishes: Adaptation to Carbonate Ingestions, Comp. Biochem. Physiol., 50A: 131-134.

APPENDIX

Concentration of ^{90}Sr, $^{239,240}Pu$, ^{241}Am in Bone and ^{137}Cs in Muscle

of Marshall Island Fish

(in pCi/kg wet weight decay corrected to Nov. 1978)

Trophic levels	Station[a]	$^{239,240}Pu$ Bone	^{241}Am Bone	^{90}Sr Bone	^{137}Cs Muscle
Group A Atolls: Ailuk (A), Jemo (S), Likiep (L), Mejit (R), Taka (H), Utirik (I), Ujelang (J), and Wotho (M)					
Trophic Level II					
Surgeonfish	A1	5.2 (15)	<0.2	22 (18)	20 (22)
	A11	1.4 (26)	0.6 (50)	20 (11)	10 (5)
	H4	4.3 (7)	0.09 (50)	17 (10)	17 (3)
	H5	12.1 (8)	0.4 (60)	21 (9)	13 (12)
	I8	12.0 (20)	1.2 (29)	<9	24 (6)
	J22	1.6 (27)	0.4 (60)	12 (46)	<6
	L50	0.9 (45)	<0.06	12 (12)	7.9 (4)
	L55	4.3 (13)		14 (12)	12 (6)
	L55	1.9 (22)	1.3 (70)	33 (65)	8 (50)
	L58	8.5 (6)	0.14 (35)	17 (8)	20 (10)
	M12				21 (7)
	M17	4.0 (10)	0.22 (33)	13 (3)	20 (5)
	S1	7.6 (3)			21 (5)
Crenimugil	A1	3.3 (8)			7.7 (23)
	A11	3.8 (5)			7.9 (8)
	A53	5.7 (8)	0.72 (34)	23 (10)	5.9 (5)
	L37	1.4 (12)	0.09 (50)	13 (8)	6.0 (2)
	L50	4.8 (6)	0.29 (15)		7.6 (3)
	L55	1.9 (12)			7.4 (19)
	M17	2.5 (11)	<0.5	<20	11.0 (11)
Neomyxus	H1	4.5 (9)	0.02 (67)	40 (5)	3.6 (10)
	H4	2.8 (9)	0.07 (50)	19 (14)	3.7 (12)
	H5	2.8 (10)			
	M1	3.1 (6)			5.8 (7)
	M12	3.6 (13)	<0.05	11 (10)	4.1 (20)
Rabbitfish	L3	0.25 (30)	<0.03	15 (4)	4.8 (10)
Rudderfish	L55	0.91 (12)	0.07 (80)	12 (9)	11.0 (3)
Unicornfish	S1	0.27 (28)	<0.01	17 (14)	8.3 (15)
Trophic Level II					
Goatfish	A11	0.13 (34)	0.04 (60)	22 (4)	6.2 (3)
	A20	<0.03	<0.05	43 (7)	5.7 (5)
	A53	<0.1	<0.1	90 (5)	7.2 (2)

APPENDIX (Continued).

Trophic levels	Station[a]	$^{239,240}Pu$ Bone	^{241}Am Bone	^{90}Sr Bone	^{137}Cs Muscle
	I1	0.04 (53)			6.2 (8)
	J5	0.13 (50)	0.15 (45)	19 (7)	6.0 (10)
	J18	<0.1		16 (8)	8.5 (15)
	L37	<0.1	<0.07	14 (12)	4.9 (5)
	L50	0.06 (60)	<0.08	20 (7)	5.6 (3)
	L58	<0.04	<0.02	14 (9)	7.2 (3)
	M1	<0.1	<0.02	24 (14)	7.0 (5)
	M7				7.2 (3)
Flagtail	I2	<0.1	<0.17 (40)	11 (12)	4.1 (10)
	R1	0.04 (50)			6.8 (10)
Threadfin	I8	<0.03	<0.05	12 (11)	15.5 (6)
	SI	0.09 (28)	0.07 (35)	7.6 (4)	13.1 (3)
Trophic Level II					
Parrotfish	I8	<0.3	0.3 (80)	12 (44)	
	L55	1.4 (21)	<0.2	8 (50)	10.0 (6)
	L58	0.42 (14)	0.1 (40)	15 (17)	14.4 (3)
	M1	1.3 (17)	0.5 (50)	10 (12)	15.2 (5)
	M12	1.1 (15)	0.9 (25)	14 (9)	13.1 (11)
Large Carnivores (III-V)					
Grouper	I	<0.01	0.16 (60)	17 (13)	24.3 (3)
	I7	0.12 (43)	0.13 (30)	13 (18)	16.6 (7)
Jack	H1	0.07 (43)	0.11 (50	14 (37)	38.0 (8)
	J5	0.66 (26)	0.09 (60)	12 (13)	15.4 (2)
Mackerel	A	<0.2	<0.1	<8	15.7 (4)
Rainbow Runner	M	<0.03		10 (25)	21.9 (3)
Snapper	H1	0.12 (50)	0.1 (70)	8.6 (7)	18.3 (3)
Jack	M	<0.04	0.15 (70)	7 (32)	17.3 (3)
Ulua	J18	<0.2	0.1 (60)	12 (9)	24.4 (2)
Group B Atolls: Ailinginae (C), Bikar (D), and Rongerik (G)					
Trophic Level II					
Surgeonfish	C5	19.6 (7)	<0.07	10 (10)	19.9 (3)
	C19	21.6 (7)	1.2 (25)	10 (49)	20.0 (5)
	C24	15.8 (5)	0.19 (35)	30 (5)	28.2 (4)
	C27	17.0 (7)	0.15 (26)	17 (5)	24.5 (2)
	D1	7.5 (11)	<1	39 (29)	22.5 (4)
	D4		<0.01	35 (4)	21.1 (3)
	G1	18.2 (4)	0.68 (16)	31 (5)	15.8 (7)
	G6	3.7 (9)	0.51 (18)	67 (7)	20 (10)

APPENDIX (Continued).

Trophic levels	Station[a]	$^{239,240}Pu$ Bone		^{241}Am Bone		^{90}Sr Bone		^{137}Cs Muscle	
	G11	4.3	(8)	0.21	(25)	35	(5)	22	(13)
	G12	8.7	(4)	0.7	(18)	38	(9)	21.2	(5)
Crenimugil	C5	13.9	(6)					13.9	(8)
	C19							11.3	(2)
	C27	16.8	(7)	1.4	(16)	27	(11)	6.5	(10)
	D1	1.28	(6)						
Neomyxus	C27	4.9	(12)	<0.08		14	(12)	5.8	(7)
	D1	1.1	(15)					6.2	(14)
	D4	2.0	(13)	0.29	(39)	29	(11)	5.7	(3)
	G1							5.3	(31)
	G11	2.5	(11)	0.46	(30)	30	(8)	7.3	(7)
Goatfish	C5	0.035	(34)	<0.01		35	(11)	7.2	(4)
	C15	<0.07		<0.01		67	(8)	6.9	(3)
	G6	0.47	(15)	0.16	(30)	135	(2)	9	(12)
Threadfin	G1	<0.03		0.2	(60)	16	(10)	16.3	(3)
Trophic Level IV									
Parrotfish	C24	7.4	(7)	0.46	(20)	25	(7)	18.1	(3)
	D1	3.3	(11)	1.2	(30)	25	(11)	24.1	(4)
	D4	1.3	(34)	0.18	(30)	20	(22)	21.5	(3)
	G11	1.6	(23)	<0.4		10	(27)	14.3	(9)
Large Carnivores III - IV									
Grouper	C	<0.06		<0.06		22	(20)	35.9	(3)
	G	0.13	(71)	<0.06		17	(23)	43.0	(9)
Mackerel	C	<0.03		<0.1		22	(24)	21.4	(2)
	G	<0.2		<0.3		19	(31)	15.7	(10)
Rainbow Runner	C	<0.09		0.28	(50)			14.3	(3)
Snapper	C	0.23	(27)	0.18	(30)	17.0	(8)	18.9	(3)
	G	0.11	(40)	0.14	(39)	16.9	(4)	15.6	(4)
Tuna	G	<0.02		<0.06		12.0	(15)	28.0	(6)
Ulua	D	<0.01		0.06	(60)	12.4	(15)	25.6	(3)
	D	<0.2		<0.06		8.7	(13)	24.8	(3)
	G	0.09	(75)	<0.09		20.0	(9)	30.8	(3)
Atoll: Rongelap (F)									
Trophic Level II									
Surgeonfish	F1	117	(4)	5.1	(6)	380	(3)	61	(10)
	F5	68	(4)	4.3	(10)	210	(2)	47	(2)
	F23	39	(5)	2.0	(20)	123	(2)	36	(9)

APPENDIX (Continued).

Trophic levels	Station[a]	$^{239,240}Pu$ Bone		^{241}Am Bone		^{90}Sr Bone		^{137}Cs Muscle	
	F33	25	(3)			78	(2)	34	(9)
	F42	11	(6)	0.47	(17)	29	(4)	17	(8)
	F46	29	(5)	1.4	(15)	43	(5)	23	(11)
	F47	28	(5)	2.4	(12)	131	(4)	24	(3)
Crenimugil	F1	35	(7)	1.4	(19)	149	(4)	27	(10)
	F5	53	(3)	7.9	(6)	466	(2)	27	(7)
	F9	38	(5)					16.2	(4)
	F23	43	(4)	0.29	(18)	212	(2)	21	(5)
	F47	16	(5)	0.28	(60)	<2		36	(6)
Neomyxus	F13	18	(7)	2.6	(11)	846	(3)	133	(2)
Goatfish	F1	0.18	(12)	0.05	(70)	52	(3)	9.5	(10)
	F9	0.74	(12)	0.13	(50)	153	(2)	7.7	(3)
	F13	1.01	(13)	0.20	(50)	351	(2)	8.7	(2)
	F33	0.34	(31)	0.19	(40)	76	(4)	7.4	(2)
	F42	0.46	(3)	0.08	(50)	123	(3)	5.8	(13)
Trophic Level IV									
Parrotfish	F5	16	(5)	0.9	(17)	24	(6)	18	(10)
Large Carnivores (III - IV)									
Bonito	F	0.06	(50)	<0.08		7.6	(11)	18.5	(6)
Bonito	F	0.17	(60)	<0.09		8.7	(14)	18.2	(2)
Mackerel	F	0.03	(50)	<0.03		4.2	(14)	27.9	(3)
Rainbow Runner	F	0.10	(80)	<0.09		12.0	(15)	14.2	(6)
Snapper	F	0.19	(29)	0.11	(50)	39.0	(4)	18.2	(3)

[a] Sample station I.D. at each atoll. Station I.D. letters without number indicates mid-lagoon locations.

[b] Value in parenthesis is the one sigma-counting error expressed as % of the concentration.

[c] Blank space indicates no analysis.